REMARKS

By this amendment, claims 1-17 have been cancelled, the title, the specification and abstract have been amended and new claims 18-33 have been added. Currently, claims 18-33 are pending in the application. These amendments have been made to place the application in better condition for the examination. Also, applicants have amended the specification to include the changes made in the parent application.

If there are any questions or concerns regarding this amendment or the remarks, the Examiner is requested to telephone the undersigned at the telephone number listed below.

Respectfully submitted,

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IN THE ABSTRACT

Please substitute the paragraphs on page 34 with the following paragraph as follows:

-- ~~Metallic materials are mixed with an organic chelating agent so as to result in a given metal composition to prepare a transparent aqueous solution of organic metal chelate complexes. The aqueous solution is spray dried to obtain an amorphous powder containing the amorphous organic metal chelate complexes. The powder is sintered to produce a metal oxide. Alternatively, the powder containing the complexes is molded into a tablet to obtain a target, which is used to form a thin metal oxide film.~~

~~The present invention provides a metal oxide whose composition and shape of the particle is easily controllable and also provide a target with which a thin metal oxide film of appropriate composition is produced effectively by a laser deposition method.~~

A process for forming a thin metal oxide film is disclosed that comprises molding an amorphous powder of organic metal chelate complexes to obtain a target. The process also includes subjecting the target to a PVD process that forms the thin metal oxide. --

AMENDMENTS TO THE SPECIFICATION

Please add the first paragraph beginning at page 1, line 6 as follows:

-- This application is a divisional application of U.S. patent application no. 09/673,920 filed on December 13, 2000, currently pending. --

Please substitute the paragraph beginning at page 1, line 2-5 to read as follows:

-- ~~PROCESS FOR PRODUCING METAL OXIDE, TARGET COMPRISING THE METAL OXIDE FOR FORMING THIN METAL OXIDE FILM, PROCESS FOR PRODUCING THE SAME, AND~~ PROCESS FOR PRODUCING THIN METAL OXIDE FILM --

Please substitute the paragraph beginning at page 1, line 7 and ending at line 12 to read as follows:

-- ~~The present invention relates to a process for producing metal oxide in which a compositional control is satisfactory, a~~

~~target with which~~ The present invention relates to a target having a satisfactorily controllable composition, with which a thin metal oxide film having a proper composition can be efficiently produced by laser deposition method, process for producing the target, and process for forming the thin metal oxide film by using the target. --

Please substitute the paragraph beginning at page 3, line 5 and ending at line 11 to read as follows:

-- A method for synthesizing ceramics from metal chelate complexes has been developed recently. This method is remarkable because of its possibility of suppressing the undesirable metal composition shift. In this method, however, an appropriate ~~mean~~ means for mixing solid-phase metal chelate complexes homogeneously at a molecular level has not been found. Thus, the method does not sufficiently make use of the advantage of the metal chelate complexes. --

Please substitute the paragraphs beginning at page 3, line 16 and ending at page 8, line 2 to read as follows:

-- On the other hand, as a method for producing a thin film of multi-element metal oxide, chemical vapor deposition (CVD) and physical vapor deposition (PVD) have been known. In CVD, a raw material is caused to react in a gaseous-phase state and the objective compound is separated out in a solid-phase state. In PVD, a solid raw material (target) is vaporized by the injection of physical energy to rearrange the vaporized material into a thin oxide film on a substrate. Of these, CVD has a drawback that a different phase is likely to be separated during the intermediate reaction due to various vapor pressures of respective kinds of raw materials. For this reason, when CVD is applied to the method of producing a multi-element metal oxide, it is considered to be difficult to control the composition of the obtained metal oxide. On the other hand, sputtering, which is a typical PVD process, is for versatile use, while it has a drawback that the compositional control is considerably difficult to be made in case of using a compound target. This is because the use of an identical compound target for a long period of time may change the composition of the target.

In order to improve the problem of the compositional control, developed has been a laser deposition method (laser abrasion) in which a target is clustered by a laser irradiation to rearrange the compounds contained in the target on a

substrate. However, strong laser energy ~~have~~ has been needed for clustering the conventional targets, since they have strong bonds such as ionic and covalent bonds. Additionally, large agglomerated molecules (droplet) of the target material may drop on the substrate, resulting in a problem that a heterogeneous thin film (i.e., a thin film having a rough surface) is obtained.

In order to solve the problem, it has been tried to provide a cloison between the target and the substrate to suppress the deposit of such a droplet on the substrate. Using the cloison, however, is only a physical ~~mean~~ means and it cannot be said a fundamental solution. In addition, the obtained thin film according to PVD is originally no better than a thin film in which the compounds in the target are rearranged. Thus, the thin oxide film quality depends on the target quality. It is therefore necessary to prepare a high-quality target. But, it is difficult to synthesize a high-quality homogeneous bulk of multi-element metal oxide to be used as the target. The synthesis also needs a lot of time and effort, and thereby the synthesis step may be the rate-determining step, resulting in a disadvantageously high cost.

The present invention is intended to overcome the aforementioned conventional problems. It is therefore an object of the invention to provide ~~a method in which, in any component~~

~~system, a metal oxide having a proper controlled composition can be easily obtained at a relatively low sintering temperature. It is another object to provide a target having homogeneity at a molecular level~~ a target having homogeneity at a molecular level in any component system. The target can be used for a speedily, low-energy and low-cost production of a high-quality thin metal oxide film, especially thin film of multi-element metal oxide, having a proper-controlled composition and a satisfactory surface smoothness. It is another object of the invention to provide a method for producing the above-mentioned target easily. A further object of the invention is to provide a method for effectively forming the above-mentioned thin metal oxide film having a proper-controlled composition with using the target.

Disclosure of Invention

According to the present invention, ~~the method for producing a metal oxide, which can achieve the aforementioned objects, has a main feature of using an amorphous powder containing organic metal chelate complexes as a raw material. The amorphous powder of organic metal chelate complexes to be used in the invention can be obtained by mixing metallic materials with an organic chelating agent so as to give a predetermined metal composition~~

~~to prepare a transparent aqueous solution of organic metal chelate complexes and then splay drying the aqueous solution.~~

~~As the organic chelating agent, preferably used may be an amino carboxylic acid chelating agent that is not thermally decomposed at 200 °C or less.~~

~~Additionally, in order to prepare the aqueous solution of the above mentioned metal chelate complexes, especially that of multi element metal chelate complexes, it is preferred that the chelating agent with at least stoichiometric quantity of a total of respective metallic material is mixed with the metallic materials so as to allow all of the metallic materials for forming complexes completely, to obtain a transparent aqueous solution. Moreover, when the metallic materials contain the metal that is easy to be oxidized by air to be a metal oxide or a higher valence metal ion, a reducing agent and/or an antioxidant may be added to the aqueous solution of the organic metal chelate complexes to prevent oxidation of the metal ions in the solution. For example, when the metallic materials contain titanium, a reducing agent may be effectively added to stabilize titanium (III).~~

~~The amorphous organic metal chelate complex according to the present invention is in an amorphous state and has a homogeneous composition at a molecular level. It also can be sintered at a~~

~~considerably lower temperature than the conventional raw materials for producing oxide films, as proved in the following practical examples.~~

~~In addition, according to the present invention, the target for forming a thin metal oxide film~~ <u>the target for forming a thin metal oxide film, which can achieve the aforementioned objects,</u> is distinctively obtained by molding an amorphous powder of organic metal chelate complexes into the shape of <u>a</u> tablet. The preferable method for producing such a target is as follows. Metallic materials are mixed with an organic chelating agent so as to give a predetermined metal composition, to prepare a transparent aqueous solution of organic metal chelate complexes. The aqueous solution is then spray-dried to obtain an amorphous powder of the organic metal chelate complexes in which the complexes are mixed with each other at a molecular level, followed by the press molding to shape into a tablet.

As the organic chelating agent ~~also~~ in this case, preferably used may be an amino-carboxylic acid chelating agent which is not thermally decomposed at 200 °C or less. In addition, the chelating agent with at least stoichiometric quantity of a total of respective metallic material is preferably mixed with the metallic materials so as to allow all of the metallic materials for forming complexes completely, to obtain a transparent aqueous

solution. Moreover, when the metallic materials contain the metal which is easy to be oxidized by air to be a metal oxide or a higher-valence metal ion, a reducing agent and/or an antioxidant may be added to the aqueous solution of the organic metal chelate complexes to prevent oxidation of the metal ions in the solution. For example, when the metallic materials contain titanium, a reducing agent may be effectively added to stabilize titanium (III). --

Please substitute the paragraph beginning at page 9, line 4 and ending at page 10, line 6 to read as follows:

-- Fig. 8 is a graph showing an X-ray diffraction spectrum of the metal oxide powder obtained in ~~practical example 3~~ reference example 1; Fig. 9 is a graph showing an X-ray diffraction spectrum of the metal oxide powder obtained in comparative ~~example 3~~ reference example 1; Fig. 10 is a graph showing an X-ray diffraction spectrum of the metal oxide powder obtained in ~~practical example 4~~ reference example 2; and Fig. 11 is a graph showing an X-ray diffraction spectrum of the metal oxide powder obtained in comparative ~~example 4~~ reference example 2.

Fig. 12 are pictures by SEM of respective powders of metal chelate complexes used in ~~practical example 4~~ reference example 2 and comparative ~~example 4~~ reference example 2 and of the metal oxides obtained by sintering the respective powders at 450 °C; Fig. 13 are pictures by SEM of the metal oxides obtained by sintering the respective powders of metal chelate complexes at 600 or 800 °C in ~~practical example 4~~ reference example 2 and comparative ~~example 4~~ reference example 2; Fig. 14 are pictures by SEM of the metal oxides obtained by sintering the respective powders of metal chelate complexes at 1000 °C in ~~practical example 4~~ reference example 2 and comparative ~~example 4~~ reference example 2.

Fig. 15 is a graph showing an x-ray diffraction spectrum of the thin metal oxide film obtained in practical ~~example 5~~ example 3.

Best Mode for Carrying out the Invention

The present invention is configured as described above. In brief, the present invention provides ~~a method in which an amorphous powder containing organic metal chelate complexes is used as a raw material for producing a metal oxide. The amorphous powder containing organic metal chelate complexes~~ a target obtained by molding an amorphous powder of organic metal

chelate complexes into a tablet. The amorphous powder of organic metal oxide complexes can produce a metal oxide by sintering at a relatively lower sintering temperature (for example, 100-250 lower) than that of the conventional method for producing an oxide as above described. --

Please substitute the paragraph beginning at page 11, line 8 and ending at line 16 to read as follows:

-- Moreover, the metal oxide powder obtained form the amorphous powder has an approximately spherical particle shape and thereby it is non-directional as describe above. Therefore, when the spherical-shaped powder is used as a starting raw material for molding, it is possible to increase the packing rate uniformly, compared with a case of using a starting material having any other particle shape. For the reason, ~~the metal oxide powder obtained by the present invention~~ this metal oxide powder may be effectively used for synthesizing high-density ceramics such as YAG (Yttrium Aluminum Garnet) and stabilized zirconia. --

Please substitute the paragraph beginning at page 11, line 24 and ending at page 12, line 6 to read as follows:

-- ~~Furthermore, when the amorphous powder containing organic metal chelate complexes is molded into a tablet to be used as a target, the metal complexes~~ According to the present invention, the target is obtained by molding the amorphous powder of the metal chelate complexes into a tablet. The metal complexes can be clustered and then easily removed from the target by the laser irradiation on the target. When the clustered metal complexes are caused to fall on a heated substrate, they are instantaneously thermally decomposed into a metal oxide so as to allow the epitaxial growth of the metal oxide on the substrate. This makes it possible to form the desired thin metal oxide film easily. --

Please substitute the paragraph beginning at page 12, line 7 and ending at line 22 to read as follows:

-- Then, a method for producing the amorphous organic metal chelate complexes is explained in detail ~~in the followings~~ below. First of all, respective metallic materials are measured so as to give a predetermined metal composition. The measured materials are caused to react with an organic chelating agent to prepare a transparent aqueous solution of organic metal chelate complexes. The reaction is performed in an aquosity medium at 20 to 100 °C, preferably 50 to 70 °C. The concentration of the aqueous

solution may be adjusted to have a solid content of 5 to 30 % by weight, preferably 10 to 20 % by weight. In addition, the amount of the organic chelating agent can be arbitrarily determined, as long as the amount exceeds the total equivalent of metallic ions. The preferable amount of the chelating agent is 1.0 to 1.5 times in mole as much as the total equivalent of metallic ions. In case that the metal chelate complexes or the organic chelating agent does not dissolve in the medium completely, ~~anmonia~~ ammonia, amine or the like can be added for the complete dissolution. --

Please substitute the paragraph beginning at page 15, line 2 and ending at line 22 to read as follows:

-- Furthermore, according to a kind of the metal, an ion of the metal may be oxidized in the aqueous solution by contacting with air to produce a metal oxide or a metal ion having a low ~~number of valances~~ valence number will change to a metal ion having a higher ~~number of valances~~ valence number. This may cause problems of solubility and stability in the aqueous solution. In a specific example of using titanium as one of metallic components, used may be an organic chelate complex of

titanium (III) having the following property. The titanium (III) ion may be oxidized in the aqueous solution by contacting with air to produce titanium (IV) that is unstable in an aqueous phase, which may be further oxidized into titanium oxide. Accordingly, in case of using such a metallic material which has the above-mentioned property, it is preferred to add a reducing agent or an antioxidant to the processing system to stabilize the metal ion in addition to inhibiting the oxidation of the metal. Then the aqueous solution of the organic metal chelate complexes is preferably prepared by mixing the chelating agent with adjusted stoichiometric quantity of the metallic materials so that complex salts of other metal ions are formed to give a completely transparent aqueous solution. The reducing agent (or an antioxidant) in this case may include ascorbic acid, isoascorbic acid, oxalic acid and hydrazine and the like. --

Please substitute the paragraph beginning at page 17, line 1 and ending at line 10 to read as follows:

-- In addition, the target produced by forming the powder of the organic metal chelate complexes into a tablet can be considerably effective as a raw material for producing a thin

metal oxide film by the laser deposition method. The method for forming the powder into a tablet may not be limited, if the obtained tablet has a strength to ~~same~~ some extent. The preferable method is a cold isotropic pressing (CIP), and the preferable compacting pressure in this method is about 200 to 1000 kg/m3. The tablet preferably has a thin cylindrical shape, from the viewpoints of the tablet strength and the cooling efficiency thereof after heating by a laser irradiation. --

Please substitute the paragraphs beginning at page 18, line 4 and ending at line 24 to read as follows:

-- ~~As described above, according to the present invention, when using the amorphous powder containing organic chelate metal complexes as the starting material for synthesizing a metal oxide, any desired form such as a powdery form, a bulk form and a thin film form of the metal oxide is easily obtainable by sintering the amorphous powder at a relatively low sintering temperature. In addition, the obtained metal oxide in these forms can have a highly controllable composition due to the use of the amorphous organic chelate metal complexes. Furthermore, the powder of chelate complexes can be shaped into any desired form such as a powdery form and a bulk form. Thus, the desired~~

~~form of the metal oxide is obtainable by pre-shaping the powder of chelate complexes prior to the sintering.~~

~~Of these metal chelate forms, in case that the powder containing chelate metal complexes is molded into a tablet to be used as a target, which is applicable to, for example, the laser deposition method~~ According to the present invention, the starting material for producing a metal oxide is obtained by molding the amorphous powder of organic chelate metal complexes into a tablet. When using the tablet as a target to be applied to, for example, the laser deposition method, a thin metal oxide film having an excellent surface smoothness can be obtained easily through an epitaxial growth with the composition of the thin film being highly controlled. This method is extremely effective for producing a high-quality thin metal oxide, especially a thin multi-element metal oxide film, which has not been produced easily by the conventional methods. --

Please substitute the paragraph beginning at page 19, line 9 and ending at line 13 to read as follows:

-- The ~~method for producing a metal oxide according to the~~ present invention is widely and effectively applied for producing a multi-element metal oxide, especially a thin film thereof, as

described above. In addition, the method is applicable to a technology for forming a single-element metal oxide and a thin film thereof. --

Please substitute the heading at page 24, line 11 to read as follows:

-- ~~Practical Example 3~~ Reference Example 1 --

Please substitute the heading at page 25, line 9 to read as follows:

-- ~~Comparative Example 3~~ Comparative Reference Example 1 --

Please substitute the paragraph beginning at page 25, line 18 and ending at page 26, line 3 to read as follows:

Figs. 8 and 9 respectively show x-ray diffraction spectra of the metal oxide powders obtained in ~~practical example 3~~ reference example 1 and comparative ~~example 3~~ reference example 1. Comparing these figures proved as follows. The powder in ~~practical example 3~~ reference example 1 has a single phase of $SrTiO_3$ at 500 °C. On the other hand, the powder at 700 °C in

comparative ~~example 3~~ reference example 1 has a phase of $SrTiO_3$ partially and, in addition, a different phase of $SrCO_2$. Furthermore, even when the sintering temperature in comparative ~~example 3~~ reference example 1 is raised up to 800 °C, the obtained powder cannot has the $SrTiO_3$ phase as a single phase.

~~Practical Example 4~~ Reference Example 2 --

Please substitute the heading at page 26, line 25 to read as follows:

-- ~~Comparative Example 4~~ Comparative Reference Example 2 --

Please substitute the paragraphs beginning at page 27, line 6 and ending at page 28, line 2 to read as follows:

-- Figs. 10 and 11 show the x-ray diffraction spectra of the metal oxide powders obtained in ~~practical example 4~~ reference example 2 and comparative ~~example 4~~ reference example 2 respectively. Additionally, Figs. 12 to 14 show SEM photographs of each powdery raw material and each powder obtained by sintering at respective temperatures in those examples.

The results shown in Figs. 10 and 11 reveals that, in ~~practical example 4~~ reference example 2, peaks of Y_2O_3 are detected even in the spectrum of the powder sintered at 450 °C in air. In this ~~practical~~ example, it is also found that the powder sintered in the oxygen atmosphere is a highly crystallized yttria. Furthermore, according to Figs. 12 to 14, the sintered powder in the ~~practical example~~ reference example 2 has a bit compressed spherical shape, resulting from the shape of its powdery raw material. On the other hand, in the comparative ~~example~~ reference example 2, only traces of Y_2O_3 peaks are observed in the spectrum of the powder sintered at 450 °C. That is, yttria is hardly produced at this temperature. In addition, this sintered powder has a directional plate crystal.

Practical Example ~~5~~ 3

An amorphous powder was obtained in the same manner as ~~practical~~ reference example ~~3~~ 1. The amorphous powder was molded into a cylindrical pellet having a diameter of 10 mm and a thickness of.5 mm by CIP at the pressure of 1000 kg/m^2, to be used as a target. --

Please substitute the paragraph beginning at page 29, line 5 and ending at line 13 to read as follows:

-- The present invention is ~~constituted as mentioned above. By applying the present method, a compositional control of the metal oxide can be easily made in any component system. In addition, the sintering can be performed efficiently and speedily at lower temperature for a shorter time. In particular, when using the target obtained by molding the organic metal chelate complexes containing powder into a tablet, it is possible to produce a high quality thin film of multi element metal oxide having an excellent surface smoothness more speedily with lower energy and cost~~ configured as described mentioned above. By using the target obtained by molding the organic metal chelate complexes-containing powder into a tablet, it is possible to produce a high-quality thin film of multi-element metal oxide having an excellent surface smoothness more speedily with lower energy and cost. In addition, in the method according to the present invention, a composition of the thin film is easily controllable in any component system. --